

Background

Phosphate (P) fertilizers are typically applied annually to agricultural fields, partly in inorganic form (e.g. $\text{Ca}(\text{H}_2\text{PO}_4)_2$). Mineral P-fertilizers contain some natural alpha-activity due to the presence of ^{238}U (among other alpha emitters). Uranium concentrations in P-bearing fertilizers have been reported to be in the range of 300 to 9200 Bq kg^{-1} of fertilizer (for both ^{238}U and ^{234}U).

The migration of U and other elements in soils depends on a large number of processes, including their interactions with other aqueous components and the solid phase (e.g., cation exchange, surface complexation) as well as time-variable water fluxes and water contents between the soil surface and the groundwater table. Predicting U transport hence requires an advanced reactive transport model integrating water flow, multiple solute transport and biogeochemical reactions. At SCK·CEN, a new reactive transport code for transient flow conditions, HP1, was recently developed. The HP1 code results from the coupling of the HYDRUS-1D water flow and solute transport model with the PHREEQC geochemical speciation model. The capabilities of the HP1 code are illustrated considering natural uranium leaching from agricultural soils to groundwater.

Objectives

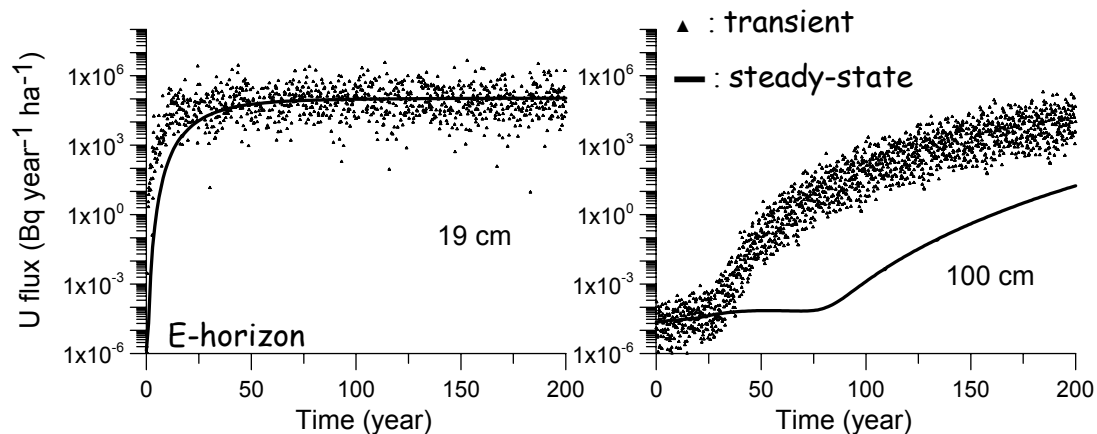
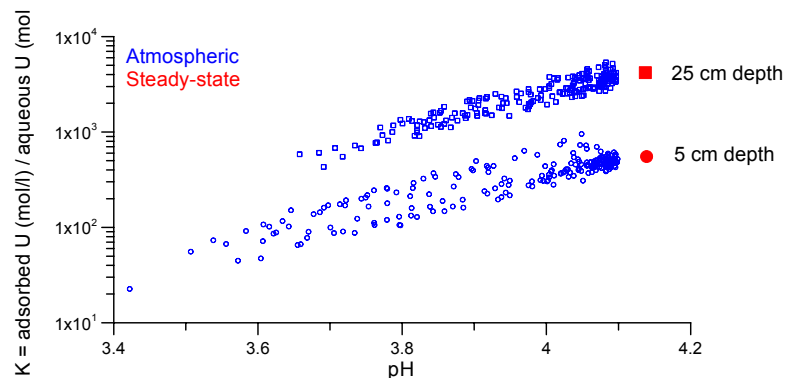
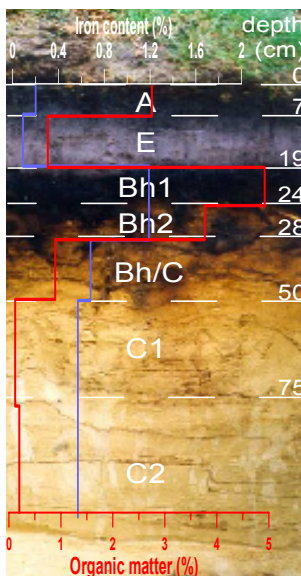
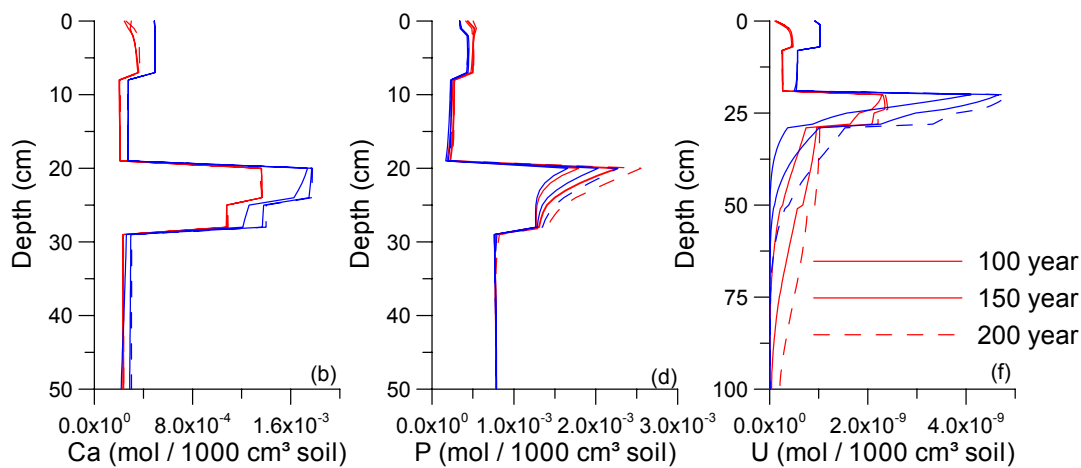
The objectives of the study are (1) to provide insight into the complex system of interacting biogeochemical processes that govern uranium mobility in soils using a new state-of-the-art coupled transport model (HP1), with special emphasis on effects of the imposed water flow boundary condition (steady-state infiltration versus atmospheric) on the migration of U in an acid sandy soil profile, and (2) to use the calculated uranium fluxes from soils to groundwater as yardsticks or reference levels for alternative or complementary safety indicators such as radionuclide fluxes from surface repositories for low- and intermediate level short-lived waste.

Principal result

The new biogeochemical transport code HP1 provides useful insights into the complex U-migration through a soil profile. Processes included in the model were non-specific binding to a multi-site cation exchange complex, specific binding on Fe-oxides, and aqueous complexation reactions. The soil (dry podsol) consists of 7 horizons with different hydraulic and geochemical properties. Photo indicates the different soil layers with the depth distribution of organic matter and Fe-oxide content indicated. Each year, we assume 0.161 mol of $\text{Ca}(\text{H}_2\text{PO}_4)_2$ fertilizer is applied containing 3.77×10^{-6} mol U. A synthetic 200-y time series of precipitation and potential evaporation represents the upper atmospheric boundary condition.

The top figure shows the depth profiles of simulated Ca, P, and U concentration after 100, 150 and 200 y. All three elements accumulate in the Bh-horizon, rich in organic matter and iron oxides (see photo). Ca is the most mobile element, followed by U, while P is mostly retained in the Bh-horizon. U reached the bottom of the soil profile after 100 year. The most important observation is that U moved faster under transient than under steady-state flow conditions. HP1 revealed that water content variations induced pH variations: when the soil is dry, the pH is lower (results not shown). The combination of the multi-site cation exchange model and the surface complexation model for competitive adsorption resulted in a smaller sorption potential at lower pH. This resulted in K -values (=adsorbed U (mol/l) / aqueous U (mol/l)) changing at least one order of magnitude (right figure second row). The lower K -values resulted in a faster U-flux at the bottom of the soil profile compared to the fluxes under steady-state flow conditions (figure last row).

By comparison, the estimated ^{238}U flux from a low-level waste surface repository (as designed in the Belgian reference concept) accounting for approximately 70,000 m^3 of conditioned waste is around $3 \text{ Bq year}^{-1} \text{ ha}^{-1}$ for an optimistic scenario when all concrete barriers are working properly for more than 10^5 years. In the case the engineered barriers are degraded (i.e., no resistance to rain water infiltrating the repository), this flux increases to $10^3 \text{ Bq year}^{-1} \text{ ha}^{-1}$. These fluxes, which would occur over several tens of thousands of years after repository closure, are comparable to the simulated fluxes from a soil profile at 100 cm depth after 150 year of continuous fertilizer application assuming transient flow conditions. However, fluxes from a repository would be much lower (up to two orders of magnitude) than "natural" U fluxes from soil when much longer time periods of fertilization would be considered (e.g., after 200 years the figure). For a surface repository, calculated radionuclide fluxes released after several hundreds of thousands to groundwater, can be used to demonstrate qualitatively that serious radiological effects due to waste disposal are not expected. Reference values may then be helpful in evaluating such fluxes from a repository. Uranium fluxes from agricultural fields could be used as potential reference values.



Future work

The present study was a first analysis in which most, but still not all, soil processes were incorporated. Further studies will expand these simulations by incorporating the effects of nitrates, carbonates (soil respiration), initial U in the soil profile, different surface sites (weak and strong sites), root U uptake, and having a more typical agricultural soil profile.

Main contact person

Diederik Jacques, diederick.jacques@sckcen.be

Main reference

Jacques, D., and Šimunek, J., 2005. *User Manual of the Multicomponent Variably-Saturated Flow and Transport Model HP1. Description, Verification, and Examples. Version 1.0.* BLG-998 Report, SCK•CEN, Mol, Belgium.

www.sckcen.be/hp1